

# Modeling Interplay Between Catalyst Performance And Microenvironment In CO<sub>2</sub> Electrolyzers

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## Abstract

With the anticipated effects of global climate change upon us, induced by egregious CO<sub>2</sub> emissions, it is imperative to implement technologies to close the carbon cycle. One potential technique to do this is storing the energy from intermittent sources (e.g. solar, wind, hydro) in the form of chemical bonds, such as the electrochemical reduction of CO<sub>2</sub> (ECR). By reducing CO<sub>2</sub> to energetically and chemically useful products of CO<sub>2</sub> (i.e. CO and C<sub>2</sub>H<sub>4</sub>) we can both reduce reliance on fossil fuels and mitigate atmospheric CO<sub>2</sub> levels. However, exerting control over the ECR is challenging because of insufficient understanding of underlying mechanisms and complexity of the overall system. First, parasitic evolution of hydrogen (HER) is always present because water serves as the proton source for ECR. Additionally, several additional variables, some not well understood, exist within the catalyst microenvironment that can also influence local CO<sub>2</sub> concentration and ECR selectivity. The observed results of research efforts exploring new catalyst materials, can be skewed due to these microenvironment conditions. For example the relatively slow diffusivity ( $\approx 2 \times 10^{-9}$  m<sup>2</sup>/s) of CO<sub>2</sub> in the liquid phase can cause inefficient utilization of catalyst active sites if turnover rates are high. Therefore, it becomes imperative to understand conditions of the catalyst microenvironment while evaluating catalysts for ECR. Studies that control parameters like catalyst layer thickness or porosity in a defined cell can provide unique insight into how external conditions can tune catalyst selectivity, activity, and stability. Utilizing COMSOL Multiphysics®, we report on the successful modeling of well-defined Ag nanowire mat catalysts assembled on a gas-diffusion support (Figure 1a-c). Simple variation to the amount of casted Ag nanowires enabled variation in catalyst layer thickness which is easily implemented in the model as well. The simulation results demonstrate that large deviations in ionic concentrations (Figure 1d) exist between varying catalyst layer thicknesses which likely induce the observed substantial differences in catalyst output (Figure 1e). Further exploration of the interplay in commercially relevant electrolyzers via modeling methods can expedite optimization catalyst architecture in the industrial sector and hopefully aid in solving the global climate crisis at hand.

## Figures used in the abstract

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**Figure 1** : (a) SEM of Ag NWs (b) Assembled Ag NWs membrane on PTFE gas-diffusion substrate. (c) Schematic of model. (d) Simulated results of K<sup>+</sup> concentration within the catalyst layer. (e) ECR performance vs Ag NW loading.